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The development and trial of an unmanned aerial system for the measurement of methane flux from landfill and greenhouse gas emission hotspots

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ABSTRACT

This paper describes the development of a new sampling and measurement method to infer methane flux using proxy measurements of CO_2 concentration and wind data recorded by Unmanned Aerial Systems (UAS). The flux method described and trialed here is appropriate to the spatial scale of landfill sites and analogous greenhouse gas emission hotspots, making it an important new method for low-cost and rapid case study quantification of fluxes from currently uncertain (but highly important) greenhouse gas sources.

We present a case study using these UAS-based measurements to derive instantaneous methane fluxes from a test landfill site in the north of England using a mass balance model tailored for UAS sampling and co-emitted CO_2 concentration as a methane-emission proxy. Methane flux (and flux uncertainty) during two trials on 27 November 2014 and 5 March 2015, were found to be 0.140 kg s^{-1} (±61% at 1σ), and 0.050 kg s^{-1} (±54% at 1σ), respectively. Uncertainty contributing to the flux was dominated by ambient variability in the background (inflow) concentration (>40%) and wind speed (>10%); with instrumental error contributing only ~1–2%. The approach described represents an important advance concerning the challenging problem of greenhouse gas hotspot flux calculation, and offers transferability to a wide range of analogous environments. This new measurement solution could add to a toolkit of approaches to better validate source-specific greenhouse emissions inventories – an important new requirement of the UNFCCC COP21 (Paris) climate change agreement.

1. Introduction

Methane is an important greenhouse gas with a 100 year global warming potential of 34 times that of carbon dioxide (IPCC: Climate Change, 2013). Landfills currently contribute around one third of the UK's methane emissions: 667 kilotonnes of methane in 2013 according to UK government databases (Defra, 2015).

The effective management and mitigation of methane emission to atmosphere from landfill gas is an important part of European Commission (EC) Guidance on landfill gas control (European Commission, 2016). However there is currently no accepted low-cost method for quantifying how much methane is emitted to the atmosphere from individual landfill sites. If the fugitive methane flux could be routinely (and cheaply) quantified, this would be a powerful tool in efforts to improve landfill gas collection rates and could serve to further minimize methane emissions from landfill sites. In addition, the method would be extremely valuable if applied for other strong emission sources such as oil and gas facilities, intensive farming and wastewater treatment.

Each of the methods currently used to make methane flux measurements from landfills has its own set of optimal and limiting characteristics in terms of the spatial and temporal scales they

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attempt to represent. Surface emissions of methane can be measured using closed chambers placed on the landfill surface. Yet in their practical limit, a lack of complete surface coverage of a site can mean that point fluxes may not be a statistically representative sample of a heterogeneously emitting site. In addition, despite their apparent simplicity, nuances in chamber design and operation, where and how they are deployed, and the use of different data analysis approaches and quality control, can have significant effects on the quality and comparability of derived fluxes. For example, a flux chamber study for a landfill in Sweden by Börjesson et al. (2000), reported a range in methane flux between −364.8 and 960 g m^{-2} day^{-1} (with uncertainties of the order 200%), noting that a site-wide flux estimate using geospatially-interpolated flux chambers disagreed with a simultaneous bulk measurement using a tracer release system, under-estimating bulk tracer flux by a factor of 4, thus noting a problem with area-normalised flux calculation when compared with point measurements and their representitiveness.

Techniques that offer coverage of a wider footprint than flux chambers carry the advantage that they can represent a measurement of an entire site or area, provided there are careful validated assumptions and appropriate modelling of atmospheric dynamics and the environmental background. These techniques include Differential Absorption Lidar – DIAL – (Innocenti et al., 2015) and tracer gas dispersion methods (e.g. Scheutz et al, 2011, Mønster et al., 2014, 2015), but these each have unique systematic (and different) limitations and constraints.

Eddy Covariance (EC) is a technique used in atmospheric science and ecology to determine exchange rates of trace gases over natural ecosystems including grasslands, forests and agricultural fields. It has been used extensively to estimate carbon dioxide and methane fluxes as part of global flux networks (e.g. Peltola et al., 2015). However, the accuracy of EC-derived fluxes can be limited by nearby steeply rising or falling topography, which can perturb the flow characteristics across the typical scale of landfill sites.

Open path Tunable Diode Laser Absorption Spectroscopy (TDLAS) is a remote sensing technique which relies on methane's strong radiative absorption in the infrared or near infrared wavelengths. Commercially-available TDLAS instruments are suitable for mounting as part of an Unmanned Aerial System (UAS) with a typical mass of ~1.5 kg, and has been used for methane measurements at landfills (Picciaia et al., 2011). However, in order to calculate a methane flux from the concentration measurements, accurate coincident wind profiles are required. However, high reported flux uncertainties (>70% relative to calculated flux) in Picciaia et al., 2011, was attributed to the absence of wind data.

Measuring an accurate whole-site fugitive methane flux from a typical landfill therefore remains technically challenging. There is currently no regulator-accepted low-cost measurement method. The absence of such a method is hindering efforts to identify and reduce methane emissions from landfills through informed and targeted regulation and responsive operational practices. UAS platforms offer the potential for a new approach to sampling and quantifying methane emission flux from landfills.

The available technology for UAS has been rapidly developing in recent years. These aerial systems now have the potential to provide sampling platforms that allow environmental monitoring to be undertaken in a way that has not been possible using ground-based instruments. The main objective of the work reported here was to develop a method for quantifying whole-site methane emissions using current UAS technology, which could then be investigated for use in the regulation of landfill sites.

In Section 2 of the paper, the approach to calculating methane flux is described together with the development of a suitable UAS methane measurement platform and instrumentation. The results of the field trial (including flux calculations) are discussed in Section 3. Forward guidance on field trial design and operating principles has been provided as Supplementary material to inform future studies.

2. Experimental methods

This section describes the methods and instrumentation used in the study. Section 2.1 describes the background to the landfill emission source. Section 2.2 describes our approach to flux calculation and Section 2.3 describes the UAS platforms and instrumentation used for the study.

2.1. Methane emission profile from landfills

Methane emissions from landfill sites comprise a wide variety of sources. These can include: permanent or temporary caps, operational (open waste) areas, waste flanks, leachate collection infrastructure, landfill gas collection systems and unburnt methane from electricity generation. A landfill can best be considered as a large, diffuse and heterogeneous area source of fugitive emissions. This complicates the assumptions often employed when attempting to derive mass fluxes from more tangible point sources of emission (such as a chimney stack for example). In addition, site emissions may also vary across different time scales due to environmental and physicochemical factors, such as temperature, pressure and soil moisture content; and site operational factors, such as the operation of the landfill gas extraction and combustion systems.

2.2. Lagrangian mass balance flux approach

The ideal sampling of a site to calculate an absolute, accurate, surface flux of any relatively inert gas (such as methane) would require complete, continuous 3D sampling with 100% measurement accuracy. Since this is simply not feasible, it is important to design and optimise a sampling method to obtain the best possible flux estimate (with a known uncertainty) within the constraints of cost, practicality and desired flux uncertainty. Emerging UAS technology may now offer a practical approach for quantifying methane fluxes relevant to the spatial scale of landfill sites in the UK (for example Illingworth et al., 2014). In a feasibility study on the use of a UAS to quantify methane fluxes, Allen et al. (2014) identified a mass balance method as a promising approach using UAS platforms.

In the Lagrangian mass balance approach, what goes into and out of a volume of air is measured with the difference in the measurements giving the net surface flux within that volume. For methane, the atmospheric lifetime is extremely long (~10 years) relative to the emission and local advection timeframe and so chemical modulation (which may be important for more reactive trace gases such as NOx) can be ignored. In this case, the total flux (in moles s^{-1}) integrated between the point of upwind and downwind measurement can be calculated from the following equation:

$$Flux = \int_0^z \int_A^B (S_{ij} - S_0) n_{ij} U_{\perp ij} \, \mathrm{dxdz} \quad (1)$$

where S_{ij} is the mole fraction (moles $mole^{-1}$) of species S for each coordinate on the vertical plane AB (oriented perpendicular to the prevailing mean wind vector), S_0 is the measured (or assumed) background, which represents the mean mole fraction upwind of the source. The n_{ij} term is the mole density of air (moles m^{-3}), which is determined using an ideal gas assumption. The $U\perp_{ij}$ term is the wind speed (m s^{-1}) perpendicular to a downwind vertical-horizontal plane AB, which spans or exceeds the lateral extent of the emitted plume. Fluxes are then integrated over the vertical

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and horizontal (AB) extent of the plane (or plume) to calculate a total flux through this plane. So long as the maximal extent of any (typically) turbulent plume is captured, and assuming that sampling time is sufficient to capture a mean state of the plume morphology, it is then possible to calculate a bulk net flux from the source. An advantage of this method is that repeated (statistical) sampling can help to constrain the natural variability in plume morphology, which would be expected due to varying atmospheric turbulence.

It should be noted that this method is fundamentally different to the Gaussian plume inversion approach, which is suited to the sampling of well-defined plumes injected above the near-surface turbulent layer from point sources. Our field experience in this study teaches us that landfills (and their many disparate and diffuse sources) cannot be conceptually modelled within the Gaussian plume framework when sampling near-to-source due to the typically diffuse and variable nature of emissions encountered on-site. The method described here therefore allows for near-source sampling, thus taking advantage of increased signal-to-noise (in terms of methane concentration enhancements over background) and therefore potentially lower precision instrumentation (reducing cost) and avoids the need to account for potential intermediate sources in the path between any source of interest and more far-field sampling necessary in Gaussian plume modelling.

Importantly, this method allows for a transparent and conservative quantification of uncertainty as the (measured) statistical variability and sources of measurement error implicit to each of the terms in Eq. (1) should be known, since each term represents a measurement from instrumentation with calibrated uncertainty and bias.

2.2.1. UAS measurements for mass balanced fluxes

The in situ gas concentration measurements made from the UAS have to be selective, fast, and provide limits of detection appropriate for ambient concentrations. The measured (or inferred) methane concentrations provide discrete data points in the downwind plane. These have to be interpolated (or extrapolated) onto a 2-dimensional regular flux grid to define a continuous measurement plane (with a known statistical uncertainty for each grid cell). This is done using the geospatial interpolation technique known as kriging (Myers, 1991). This approach has to consider: the measurement error at each sample point; the correlation length - how strongly to weight, with distance, the extrapolated concentration in the null space to its nearest neighbour sample locations; and the limiting variance between pairs of measurements as the distance separating them becomes large. These factors need to be defined in a way that best represents knowledge about the system and sampling under investigation in order to minimize uncertainty. In summary, the key to optimal and successful (and operationally useful) kriging relies on dense and rapid sampling of the plume. The sampling rate and density of measurement coverage for a site must therefore be balanced against the expected temporal rate of change of flux and any potential change in background inflow (e.g. from extraneous sources of methane upwind). Small UAS platforms can provide the required rapid dense sampling of air over a typical scale of landfill site.

2.2.2. Downwind methane measurements

Rather than measuring upwind and downwind of the source, it can be possible to use downwind measurements outside of the landfill plume as background concentrations, provided that spatial variability in the background can be expected to be low. In this context, optimal conditions for this method can be conceptualized to be free-flowing (laminar) air with consistent prevailing winds (i.e. no gusting) at either the near-surface, or in the lowermost 120 m profile, whereas poor conditions might reflect stagnant or turbulent flow or strong vertical sheer. This downwind-only approach maximizes the UAS flight time spent sampling the plume of interest and therefore facilitates better downwind statistics. The methane concentration (above the background value found outside of the plume) can be measured throughout the vertical plane downwind of the landfill and the amount of methane passing through this plane per unit time can then be calculated from the kriged (spatially interpolated) concentration and wind data. When a mean background is subtracted, the resulting flux is then equal to the net bulk mass of methane emitted per unit time by the entire landfill surface area. This is true provided that the lateral and vertical extents of the sampling in the downwind plane are designed such that it exceeds the expected dimensions of the evolved landfill plume.

2.2.3. Wind measurement

The accurate measurement of wind (and wind variability) is essential for the calculation of fluxes (and flux uncertainty) from a UAS using the mass balance approach described earlier. Ground-based meteorological sensors may not provide the necessary spatial and vertical resolution and winds should ideally be measured onboard the UAS. However, onboard measurement of wind was trialled using a 5-hole probe (see Table 1 for details) but performed poorly when compared to static sonic anemometers on the ground. Therefore, for this study, a 20 Hz sonic anemometer positioned on a 2 m tower at the launch location was used to derive a mean wind speed and direction (and wind speed and direction variability, derived as a 1-standard-deviation statistic). These data were then used as factors in Eq. (1) to derive flux (and flux uncertainty by error propagation). For interest, we refer the reader tovan den Kroonenberg, 2008, for further details of the 5-hole probe, which we hope to further develop for UAS use in future work.

2.3. UAS platforms and instrumentation

The best approach to quantifying methane flux at landfill sites in this study was considered to be a fixed-wing UAS incorporating in situ measurements of CO_2 concentrations and wind (Allen et al., 2014), complemented by rotary UAS vertical profiles of both CO_2 and CH_4 concentrations. This Section describes two UAS platforms developed and used in this case study and their respective instrumentation. In addition, the Supplementary material to this paper offers operational and logistical guidance for future studies and further details the specific design and rationale for the field trial in our case study.

At the time of the project, there was no high precision methane instrument (defined nominally here as >100 ppb at 1σ @ 1 Hz) suitable to fly on a small fixed-wing UAS – though such instruments are known to be in development. In response to this, the following approach was adopted: measurements of CO_2 concentration were recorded across the landfill gas plume downwind; a representative ratio of co-emitted carbon dioxide to methane landfill gas emissions was established (noting that landfills typically emit CH_4 and CO_2 in a specific ratio at any given time). Methane concentrations in the landfill plume were then inferred using the measured site-specific carbon-dioxide-to-methane emission ratio.

In order to achieve this, two UAS platforms were developed for the study. The first consisted of a fixed-wing platform with an onboard high-precision carbon dioxide infrared reference cell. The second consisted of a rotary hexrotor platform with a tethered 100 m-length PFA sampling line (weighing 35 g/m) to sample vertical profiles of both methane and carbon dioxide using high precision ground instrumentation (otherwise too heavy to fly). The

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Table 1
UAS platform specifications. The masses of each component are given in parentheses.

UAS	Instrumentation
Fixed-wing	5-hole probe for three-dimensional wind profiles (98 g) Edinburgh Instruments Gascard NG II carbon dioxide sensor (~1.0 kg) PIXHAWK flight control system (FCS) unit, running ArduPlane firmware version 3.2.0 (100 g) Pitot-static probe for estimating airspeed for the FCS unit (15 g) Beaglebone Black micro-controlling unit running Arch Linux for fusing information from the carbon dioxide sensor, 5-hole probe and FCS unit (120 g) 433 MHz radio telemetry to oversee real-time progress on the mission (via ground station software) and of the sensors (via a terminal window) (13 g)
Rotary	IST AG HYT-271 humidity and temperature sensor (5 g) 100 m Air sampling Teflon tube attached to a Los Gatos Research Ultraportable Greenhouse Gas Analyzer (LGR-UGGA) on the ground (35 g/m) PIXHAWK flight control system, running PX4 Copter (100 g) Radio telemetry to oversee real-time progress of the mission via ground station software (15 g)

specifications of the two UAS platforms and their instrumentation are summarized in Table 1.

While tethered-tubing systems may permit the use of more accurate ground-based instrumentation, this presents logistical challenges such as kinking and snaring on surface objects and lateral sampling may be limited by the length of tube used. Free-flying UAS offer more maneuverability but require the measurement payload to be onboard, which typically represents a weight constraint for current precision CH_4 instrumentation. The dual carbon dioxide and methane approach (referred to herein as the *proxy method*) may not be required in future work when suitably precise (and light-weight) methane instruments become available for UAS use, and that tether use may also not be required.

The fixed-wing platform used in this study was the Bormatec Explorer, a twin motor electrically powered aircraft with a maximum take-off weight of around 7 kg (this small size means that a road or runway is not required for launching and recovery). The combination of payload and battery provides sampling durations of between 20 and 30 min with a typical cruise speed of approximately 15 ms^{-1}. This allows the sampling of a reasonable plume area downwind in a single flight.

A computer running Linux Ubuntu was used as the ground station to oversee in real-time the behaviour of the flight control unit and to trigger fail-safe mechanisms in the event of radio control loss.

The simultaneous CO_2 and CH_4 in situ gas measurements for the rotary UAS were recorded using a Los Gatos Research Ultraportable Greenhouse Gas Analyzer (LGR-UGGA). This instrument uses the principle of off-axis Integrated Cavity Output Spectroscopy (OA-ICOS, see Baer et al., 2002, for details) and two near-infrared tunable diode lasers, which rapidly scan a single strong (and isolated) absorption line of the target gas. The instrument also measures water vapour concentration to accurately correct for water vapour dilution and absorption line broadening effects, to retrieve ambient CO_2 and CH_4 concentrations as a dry mole fraction without drying or post processing of absorption spectra. The measurement rate was selected at 1 Hz with a nominal 1σ Allan variance of 1.08 ppb and 0.22 ppm, for CH_4 and CO_2, respectively, characterised by sampling certified standards in the laboratory. The measurement range for the LGR-UGGA is 0.01–100 ppm and 200–2000 ppm, for CH_4 and CO_2, respectively. We also characterised the UGGA in the laboratory using WMO-traceable reference standards, certified by EMPA. All measurements by the UGGA when measuring cylinder air fell within 1% of the certified concentration; therefore, as a conservative measure, we use an arbitrary maximal uncertainty of 1% relative concentration to propagate flux uncertainty. The power requirements of the LGR-UGGA are between 60 and 66 W depending on operating procedures. Further technical information on the LGR-UGGA can be found in Allen et al., 2015.

The CO_2 fixed-wing measurements were made using an Edinburgh Sensors Gascard® NG. This utilizes non-dispersive infrared (NDIR) sensing technology, where an optical filter is used to select a narrow wavelength band from an infrared light source before it is directed through a gas cell containing the sample. By analysing the difference between the signal generated at the wavelength of an absorption line of the species of interest and the signal generated at a nearby wavelength separated from the absorption line, instrumental factors can be accounted for and the mole fraction of the target species can be derived. The model used weighs 0.3 kg and has a measurement range of 0 to 2000 parts per million (ppm) CO_2, sampling at 1 Hz. This instrument was compared to the LGR-UGGA in a side-by-side measurement prior to (and after) flight to ensure comparability within the 1% uncertainty constraint.

The measurement of wind speed and direction is more challenging using multi-rotor aircraft as the measurements need to be taken outside of the sphere of aerodynamic influence of the UAS propellers, hence our decision to use ground-based anemometers in this case study.

The rotary wing platform was a modified DJI F550 hex rotor aircraft (with a take-off weight of approximately 600 g), custom designed for atmospheric sampling. The UAS was powered through a tether around 100 m long (using a 2 kW petrol generator positioned downwind so as not to contaminate the measurement), which included a Teflon sample tube. A typical field sampling flight in this study consisted of 10 min of sampling at 10 m vertical intervals from 10 m to 100 m above local ground level. The lag time of flow through the tubing from the sample point aloft to the UGGA on the ground was measured (using a cough test to spike CO_2) to be 174 s.

3. Results and discussion

Two landfill sites operated by a single company in England were selected for the field trial. The precise locations of these sites are not given in this paper for reasons of commercial sensitivity. However the location is irrelevant to the scope of this paper, which serves to demonstrate the concept, design and test of a new UAS-based flux method. However, it should be noted that both sites were typical of UK landfills, representing large sites containing both historic (capped) and active (filling) cells. One site was situated in the north of England (referred to herein as Site 1) and the other in the east of England (referred to as Site 2). Fluxes are reported in Section 3.2 for Site 1 only as only concentration measurements (not winds) were sampled at Site 2, which precludes a flux calculation. However, we include a discussion of the concentration measurements at Site 2 in Section 3.1 below for the purposes of evaluating the efficacy of the proxy method.

Two successful fixed-wing UAS flying operations were conducted on 27 November 2014 and on 5 March 2015 at Site 1. In addition, three hexrotor flights were conducted in August 2014 at Site 2 as part of the Natural Environment Research Council's Greenhouse Gas UK and Global Emissions (GAUGE) project.

3.1. Measurements and sampling

Concentrations of carbon dioxide were measured using the fixed-wing UAS during the two flights along with GPS telemetry. Figs. 1 and 2 illustrate the horizontal flight pattern (from above)

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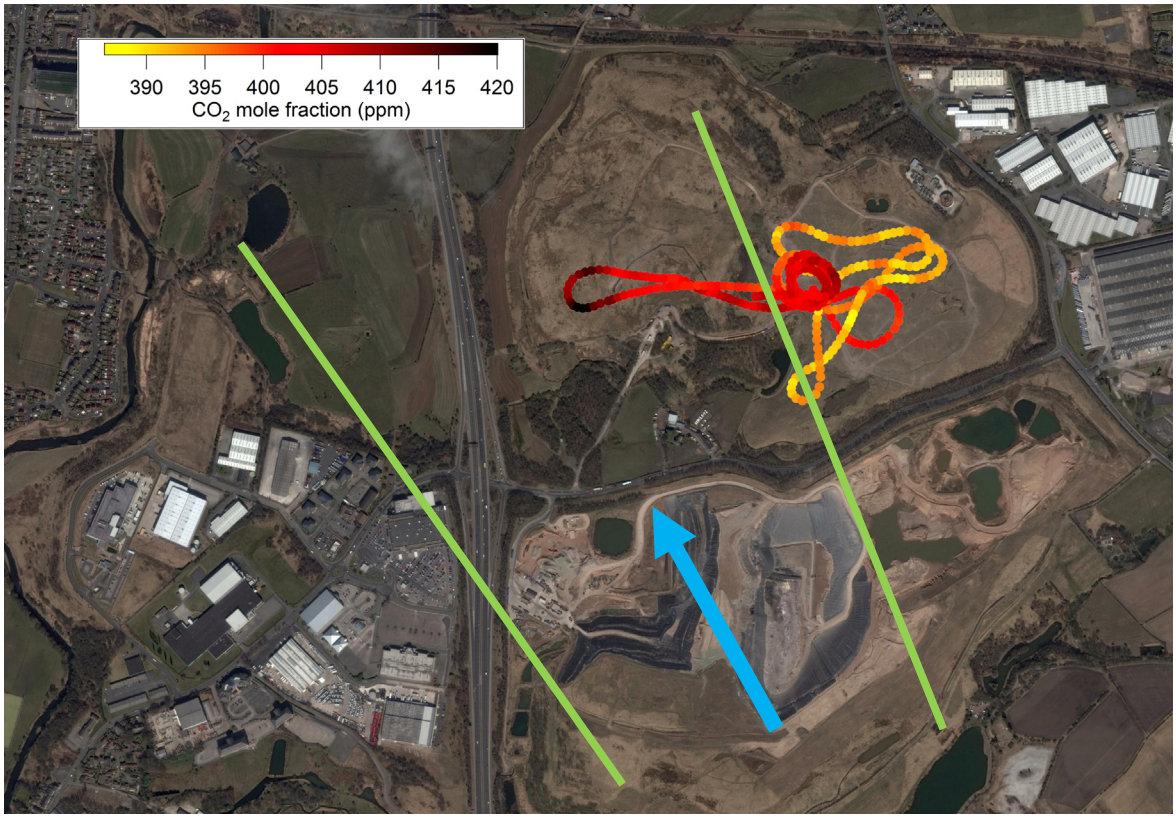

Fig. 1. Flight track and CO_2 concentrations (scaled to color legend) on 27 November 2014 at Site 1. Blue arrow shows the average wind direction on the day and green lines illustrate the expected landfill plume extent. Map data: Google, <http://www.earth.google.com>, Digital Globe, 2012 [accessed 12 March 2016]. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

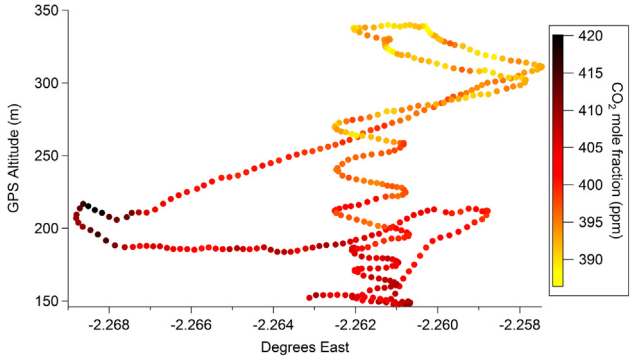

Fig. 2. Vertically-profiled measurements (referenced to longitude) of CO_2 concentration (color-scaled as per legend) by the fixed-wing UAS on 27 November 2014 at Site 1.

and the vertical-longitudinal profile of carbon dioxide concentrations on 27 November 2014, respectively. Figs. 3 and 4 show data for the flight on 5 March 2015, with the exception that Fig. 4 illustrates a vertical-latitudinal profile.

Fig. 1 shows an increasing horizontal CO_2 gas concentration gradient as the UAS penetrates the eastern extent of the plume. On this day the take-off area to the north of Site 1 was within the extent of the plume due to southeasterly winds with more background CO_2 concentrations of ~390 ppm seen to the east of the take-off location. Fig. 2 illustrates a decreasing vertical concentration gradient with height within the plume up to around 250 m altitude (~100 m above ground level), where concentrations return to background conditions suggesting a plume rise height of around 100 m at the point of sampling to the north; noting that the take-off point is 510 m to the north of the center of the open landfill. Due to the presence of a motorway to the west of Site 1, it was not possible to extend the UAS sampling further west than that shown in Fig. 1 due to Civil Aviation Authority (CAP 722 and CAP 393, see Civil Aviation Authority, 2012 and Civil Aviation Authority, 2015) regulations.

Fig. 3 (March 2015) shows a much more homogenous concentration field than that seen in Fig. 1 (November 2014), with small but measurable enhancements in CO_2 concentration sampled everywhere in the downwind environment, reflective of the fact that all sampling was generally within the trial site plume for winds blowing from the southwest on this day. However, strong enhancements such as those seen in November 2014 (up to 420 ppm) were not observed on this day and no clear plume top was observed. This creates a challenge for flux calculation using the mass balancing method, which will be discussed in Sections 3.2 and 3.3.

Concentrations of carbon dioxide and methane were also measured simultaneously using data sampled at 2 m above local ground level (using the Los Gatos Research ultraportable greenhouse gas analyzer - LGR-UGGA) at the location where the fixed-wing UAS was launched. Those measurements were used to derive representative carbon dioxide and methane emission ratios (see Figs. 5 and 6). This proxy method was tested to derive methane concentrations using linear and scalable relationships with measured carbon dioxide concentrations. This was found to be predictable and potentially useful at sites such as the one used in the GAUGE project, where it can be assumed that there are no significant nearby offsite sources of methane and carbon dioxide. However, the method was found to be subject to much higher

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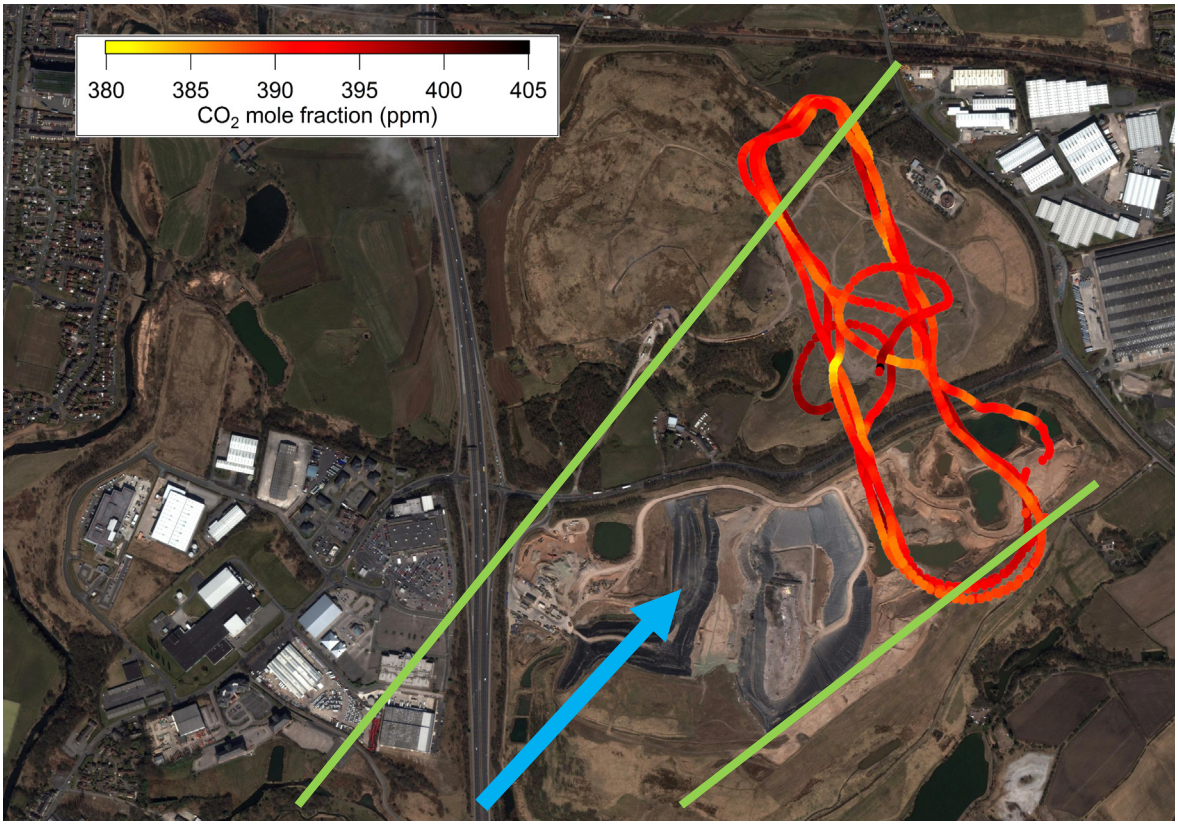

Fig. 3. Flight track and CO_2 concentrations (scaled to color legend) on 5 March 2015 at Site 1. A blue arrow shows mean wind direction on the day and green lines illustrate the expected landfill plume extent. Map data: Google, <http://www.earth.google.com>, DigitalGlobe, 2012 [accessed 12 March 2016]. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

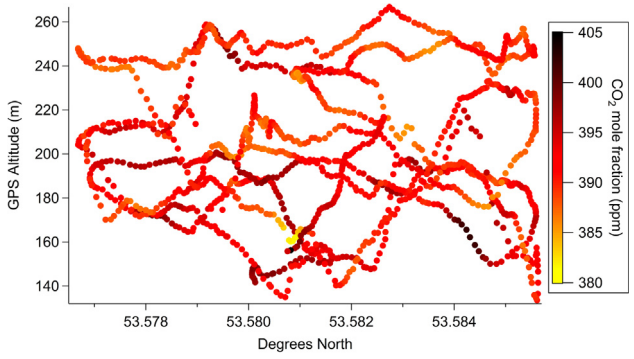

Fig. 4. Vertically-profiled measurements (referenced to latitude) of CO_2 concentration by the fixed-wing UAS on 5 March 2015 at Site 1.

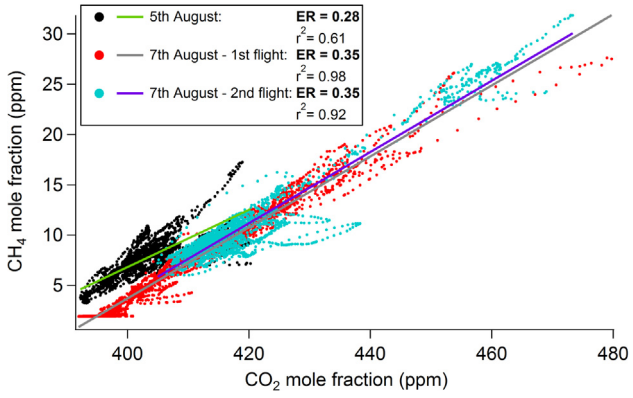

Fig. 5. Scatter of simultaneous CO_2 and CH_4 measurements at Site 2 for 3 flights (as colour-coded). Goodness of fit statistics are shown in the upper left inset.

uncertainty when practiced at sites within a few km downwind of sources such as motorways, cities or other strong greenhouse gas emitters, which affect the stability of the proxy emission ratio used as CO_2 concentrations may be more variable. This can be seen in Figs. 5 and 6, which show that there is a relatively robust characteristic emission ratio (with an R^2 of up to 0.9) for Site 2 (Fig. 5), a more rural site, but a much more variable relationship at Site 1 in November 2014 (near to an urban and industrialized area), with $R^2 = 0.36$, which can introduce a significant source of additional uncertainty when using such a variable emission ratio as a proxy for methane. The tracer-tracer relationship for the flight in November 2014 (top panel of Fig. 6) is also clearly highly variable and we see the presence of multiple linear mixing lines with variable CO_2 offsets. This reflects the variability in the CO_2 background superimposed on the characteristic (landfill-specific) gradient. Since it is the gradient (and not the offset) that is important in defining a ratio in the proxy method used here, we distil a representative ratio from the gradient of a singular mixing line for the November case (using the blue data in the top panel of Fig. 6) and retain the observed variability in the CO_2 offset as an uncertainty term that can be propagated through Eq. (1) to place a representative statistical uncertainty on the calculated flux using such variable data. Comparing the emission ratios, we clearly see a large difference between the November and March flights (0.94 and 0.3, respectively). This is interesting as it suggests a change in the relative emission from the surface of the landfill, perhaps illustrating

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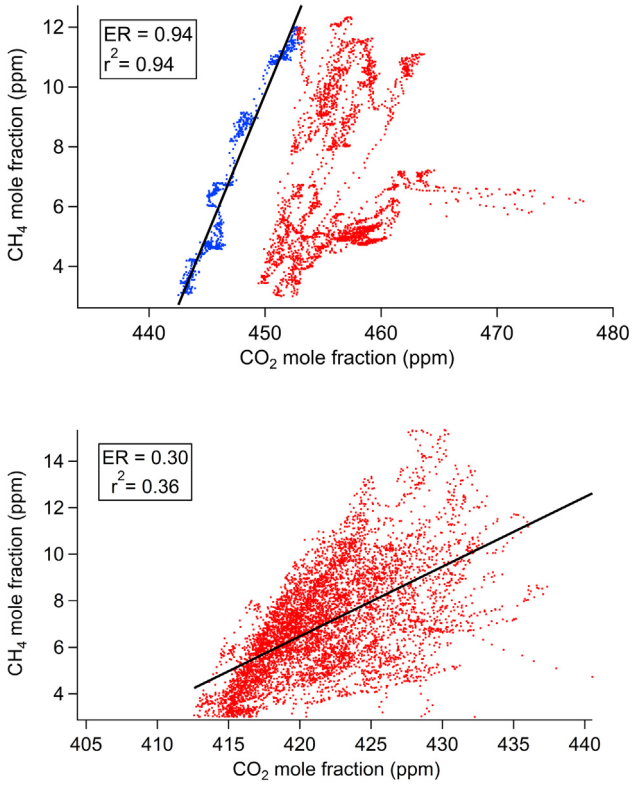

Fig. 6. Scatter of simultaneous CO_2 and CH_4 measurements using LGR-UGGA data recorded on the ground, for: top panel) - 27 November 2014; and bottom panel)- 5 March 2015 at Site 1. Confidence of fit statistics are also shown (upper left for each panel).

near-surface microbial oxidation. However, we lack further data to investigate this further and is beyond the scope of this work.

A methane-only measurement on a fixed-wing UAS will clearly have advantages over the system described here due to bypassing the proxy step and the potential uncertainty that this introduces. However, background variability in methane due to other sources must still be considered when siting measurements and planning sampling. In all cases, the upwind or out-of-plume environment needs to be sampled to assess any background variability that must be accounted for in the flux uncertainty implicit to Eq. (1).

The nature of the sampling plotted in Fig. 1 demonstrates that measured data are sampled at variable distances downwind of the landfill perimeter. Therefore, these data were projected onto a 2-dimensional flux plane perpendicular to the mean wind vector (as shown in Fig. 2). The optimal requirement that the width of the flight track (along the prevailing wind vector) is small relative to the distance downwind was, in practice, a compromise between accessible locations to fly downwind and the minimum turning circle of the fixed-wing UAS. The sampled data was interpolated on a 2D grid, spanning the vertical and horizontal extent of the plume using the kriging geospatial interpolation technique with Gaussian covariance on a 25×25 grid (as described in Section 2). The spatially-interpolated methane enhancement projected onto the flux planes are shown in Fig. 7 for both UAS flights at Site 1, after conversion from CO_2 to CH_4 using the relevant proxy ratio and subtraction of a mean background, with heights referenced to ground level of the launch site. Note also that range (x-axes in Fig. 7) were first derived relative to the radial between the mean wind vector and flux plane and converted to distance on a Great Circle using GPS coordinates. Cells within the flux domain where there was no UAS sampling were set to zero (shown as white or null space in Fig. 7) to avoid edge effects that could bias the total flux, and their surface area is then discarded within the flux plane when summing fluxes across the plane to calculate total flux using Eq. (1). Fig. 7 shows sampling of a clearly defined methane plume within the sampled domain of both flights. Ideally, the entire flux plane would be fully (and repeatedly) sampled to ensure that variable plume morphology was completely characterised; however, by not including under-sampled areas in the flux calculation, we can be confident that the flux calculated is internally consistent. Future considerations of this method should include an evaluation of the length and scope of sampling necessary to yield confidence thresholds when sampling a known source flux and will form part of our future work.

Having obtained the spatially interpolated flux planes downwind (with the mean background subtracted to represent net downwind enhancement) in Fig. 7, Eq. (1) was used to calculate a methane flux by integrating across the horizontal and vertical extents of the 2D sampling plane perpendicular to the prevailing wind (as described in Section 2.2.1).

3.2. Flux results and discussion

Methane fluxes (and concomitant uncertainty) have been calculated from the field trial data for the two UAS flights conducted at Site 1. Note again that fluxes were not derived for initial test data collected at Site 2 as winds were not sampled at that location. These are presented in Table 2 as an illustration of the method in this case study.

For the flight on 27 November 2014, the mean site-total methane flux was found to be 0.140 kg s^{-1} (±61% at 1σ). Table 2 shows that this statistical uncertainty on the derived flux is dominated by the background variability; accounting for 83% of the total flux uncertainty, which represents 59% uncertainty relative to the mean flux itself. For the flight on 5 March 2015, the flux was derived to be 0.0504 kg s^{-1} (±54% at 1σ) with an uncertainty dominated on this day by variability in both the background variability (accounting for ~51% of the total flux uncertainty) and the wind measurement variability (accounting for ~19%).

There are two reasons for this relatively large uncertainty in the case of the sampled data obtained in these two trials: (1) The calculation of the background concentration (and background variability) was derived from downwind data (due to siting and flying constraints); (2) It was not possible to confirm that the assumed background sampling was not partially contaminated by the plume.

This background was derived from a relatively uncertain emission ratio (Fig. 6), noting that the 1σ residual of the proxy emission ratio coefficient has been included in the uncertainty budget for the proxy method used here (see Section 3.3). In future, these two uncertainty components may be more tightly constrained by sampling upwind of the site to gain better confidence on the background, perhaps using multiple simultaneously flying UAS and by using a precision methane sensor onboard the fixed-wing UAS to avoid the uncertainty that results from the proxy method, where nearby sources of carbon dioxide may affect the linearity of the emission ratio.

For the flight on 5 March 2015, the mean Site 1 net methane flux was calculated to be 0.0504 kg s^{-1}, with a 1σ uncertainty of 0.0272 kg s^{-1}, again dominated by the background uncertainty for the same reasons as discussed for the 27 November 2014 case. The slightly lower relative uncertainty on this day (51% relative to the mean flux) is attributed to the more dense downwind sampling compared with that possible on 27 November 2014.

The uncapped landfill surface area of the trial site was approximately 20,000 m^2 (though it should be noted that this can vary on a daily basis). Equating the fluxes to a surface-area normalised flux per day (as sometimes reported in surface-based flux studies of landfill), the fluxes measured on 27 November and 5 March are

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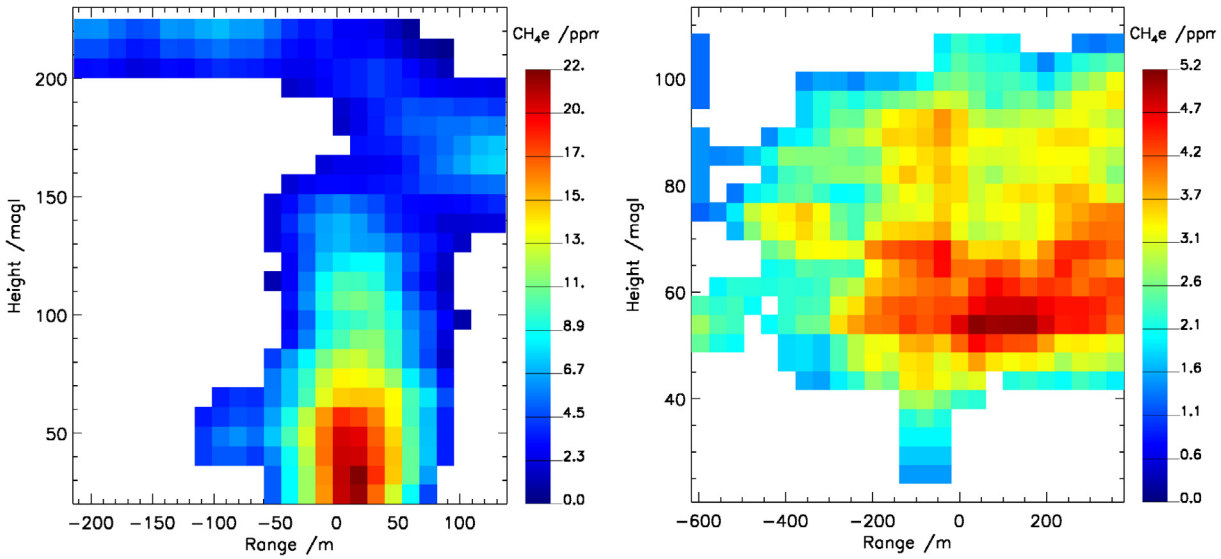

Fig. 7. Methane enhancement (CH4e) over background (in units of ppm as per colour scale) spatially interpolated onto a 2D flux plane for fixed-wing UAS flights on: left panel) 27 November 2014 at Site 1; and right panel) 5 March 2015 at Site 1.

Table 2
Mean methane flux and one standard deviation uncertainties for the two trial flight days at Site 1. All units are in kg s^{-1} rounded to 4 d.p.

Units in kg s^{-1}	Date of flight	
	27 November 2014	5 March 2015
Mean flux	0.1400	0.0504
Total uncertainty	0.0854	0.0272
Wind uncertainty	0.0018	0.0051
Background uncertainty	0.0698	0.0139
Measurement uncertainty	0.0030	0.0011
Downwind uncertainty	0.0120	0.0071

604.7 g m^{-2} day^{-1} (to 4 s.f, or 37.81 mol m^{-2} day^{-1}) and 217.7 g m^{-2} day^{-1} (to 4 s.f, or 13.61 mol m^{-2} day^{-1}), respectively. It should be noted that this calculation assumes that all emissions are associated with the uncapped landfill area, which may not be the case for reasons discussed in Section 1, highlighting a wider problem with area-normalised flux reporting such as those employed conventionally. A bulk whole-site flux is much more instructive for accounting purposes but this then provides a quantity that is less easily comparable between sites of different surface area.

The fluxes (and flux uncertainties) calculated for Site 1 are broadly comparable in absolute magnitude with those reported for analagous solid waste landfill in published literature when comparing to other whole-site emissions calculation methods. For example, a recent synthesis study of DIAL fluxes measured for both open (uncapped) and closed (capped) landfills by Innocenti et al. (2017), states that fluxes observed over a range of sites (and for several measuerments per site) are significantly higher for active sites (192–720 kg/h, or 0.0533–0.2000 kg/s) than closed sites (21–146 kg/h, or 0.0058–0.0406 kg/s). Since Site 1 measurments in this study were dominated by flux from an open landfill in the upwind footprint, and comparing the DIAL fluxes above with results in Table 2, we see that our data fits entirely within the range reported for several other UK landfill studied by Innocenti et al. (2017), using DIAL. Comparing flux uncertianty between these methods, Innocenti et al. (2017) suggest a range of 20–30% total relative flux uncertainty for a single DIAL measuerment, compared with 51–61% for a single UAS flight here. This is a very useful comparison, as we have highlighted many ways in which UAS-derived flux uncertainty may be reduced further, e.g. use of an onboard CH4 sensor, longer UAS sampling times, better wind measurements etc. Furthermore, UAS flights offer the potential to be far cheaper as a survey method than large and complex DIAL systems. Therefore, comparable flux accuracies may be possible using UAS at a fraction of the cost of DIAL systems in future.

We can also compare our whole-site fluxes with fluxes derived from tracer release experiments. Scheutz et al. (2011), performed three field campaigns at a landfill in Denmark, to study old and new landfill cells and on-site facilties. Scheutz et al. (2011), reported that the average CH_4 emissions from an old landfill section were 32.6 ± 7.4 kg CH_4 h^{-1} (or 0.0091 kg/s) and 10.3 ± 5.3 kg CH_4 h^{-1} (0.0029 kg/s) from a new section. This is a much smaller flux than that measuerd for Site 1 in our study but a direct comparison between individual sites may not be meaningful (ranges may offer a better comaprison). However, comapring flux uncertainties for the tracer release and UAS methods, we see that the tracer release experiements in Scheutz et al. (2011), suggest a range of 22–51.5%, which is similar to that using a proxy method here for single UAS flights, sampling for around 15 min. The cause of the differences in flux between our November 2014 and March 2015 UAS flights at Site 1 cannot be deconvolved as we lack the underpinning biogeophysical process data that would be needed to examine changes in microbial chemistry. However, the comparison between the two UAS measurements at Site 1 and their comparison with other whole-site approaches simply offers three instructive points: first, that the uncertainties associated with bulk techniques such as mass balancing (of the order 50% in this study) and tracer release methods can be an order of magnitude better (in terms of uncertianty) than geopatially interpolated techniques such as flux chambers (as discussed in our Introduction here); second that fluxes from landfill generally can vary widely (due to a range of local environmental factors), and finally that area-extrapolated fluxes may not be a useful measure of total emission flux when attempting to account for site-wide emissions or in understanding the physical and chemical processes governing methane generation and emission on any individual site. In summary, understanding the physico-chemical processes generating methane may unavoidably require very different mesasurement approaches to those seeking to simply measure an accurate bulk site flux. For example, measurements of soil chemistry and biology, as well as waste chemistry and environmental controls would all be required to lead to any meaningful understanding that links flux to biogeophysical processes.

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3.3. Flux uncertainty considerations and calculation

Eq. (1) was also used as an error propagation model to calculate the upper and lower bounds on the methane flux. This was achieved by quantifying the statistical uncertainty in each term of the mass balance flux equation (Eq. (1)); for example: concentration measurement (instrument) precision, sampling bias (kriging uncertainty), vertical mixing height knowledge and dilution, measured wind variability and background concentration statistical variability (i.e. the Gaussian statistics of measured wind and background concentrations during the flight). The latter term also convolves knowledge of the statistical confidence in the emission ratio used if a proxy method has been used (as was the case here). It is the variable nature of the emission ratio (due to background variability in CO_2) in this field trial that dominated this error term for Site 1, which was close to sources of extraneous CO_2 emission (several motorways and a large city within 15 km upwind).

When using high-precision, calibrated instrumentation such as that used in this field trial, it was observed that measurement (instrumental) error accounted for only ~2% relative to the derived flux. More constrained flux calculations may be obtained by avoiding the proxy step as discussed earlier, and on choosing flight days with moderate and invariant wind speeds – not too slack so as to be dominated by local turbulence, and not too strong so as to compromise flight safety. Ideally, a site-specific range of wind speeds that might be expected to give relatively laminar flow over the flight domain could be calculated using computational fluid dynamical (CFD) models. However, this is highly complex and requires a 3D model of the site topography. However, a range of wind-speeds between 2 and 10 ms^{-1} may be considered useful generally; and regardless of predicted flow characteristics, wind variability can be captured in any error budget using the above error propagation method if sampled well to gain good statistics.

The flux uncertainty arising from the sensitivity to various error components can easily be propagated in advance, synthetically, through Eq. (1) when designing a sampling system to meet a nominal error constraint. For example, using synthetic data, a 100 ppb methane measurement uncertainty would result in only around a 1% uncertainty on a flux such as that calculated above. So long as measurement uncertainty is a random Gaussian error (as opposed to systematic instrumental drift), then this error reduces proportionally with the square root of the number of measurements. This is important, as it suggests that in situ instruments with much lower point accuracy (but known random error profiles) and high sampling rates (for example, 1 Hz) can yield meaningful measurements for flux calculation; provided that dense sampling is performed in the downwind environment.

This uncertainty analysis suggests that a methane sensor with an accuracy of ~100 ppb (1σ @ 1 Hz) could be capable of delivering meaningful results (defined as <50% 1σ uncertainty for analogously derived bulk flux) using the mass balance method for landfill sites of similar size and emission profiles.

Finally, the developed method demonstrated here offers potential as a relatively low cost approach to quantifying the methane released from landfills and other greenhouse gas emission sources on a case study basis. As such, this measurement solution could add to a toolkit of approaches to better validate source-specific greenhouse emissions inventories – an important new requirement of the UNFCCC COP21 (Paris) climate change agreement.

4. Conclusions

We have described the development of a new sampling and measurement method to infer methane flux by mass balancing using proxy measurements of CO_2 concentration recorded by Unmanned Aerial Systems (UAS). The UAS flux approach is demonstrated as a cheap and novel alternative to other whole-site measurement techniques such as DIAL and tracer-release experiments with similar inherent uncertainties (~50% relative to mean flux), which may be improved further using onboard CH_4 precision sensors (as sensor technology advances), onboard wind measurement and greater sampling time (to better characterize mean plume morphology).

The approach here is appropriate to measurements of fluxes at the spatial scale of landfill sites and other strong greenhouse gas emitters such as oil and gas facilities and wastewater treatment plants. The method also benefits from its maneuverability and 3D sampling and ability to sample in the near-field (closer to sources than is often necessary in Gaussian plume inversion.

Two test flights were conducted to evaluate flux calculation for a landfill in North West England. Derived methane fluxes (and flux uncertainties) during two trials on 27 November 2014 and 5 March 2015, were found to be 0.140 kg s^{-1} (±61% at 1σ), and 0.050 kg s^{-1} (±54% at 1σ), respectively.

This work represents an important advance concerning the challenging problem of greenhouse gas hotspot flux calculation, and offers transferability to a wide range of analogous environments, including the measurement of fluxes of any other trace gas (or aerosol), where UAS-installed instrumentation may be available, and may therefore open up exciting new pathways in atmospheric process understanding.

This new measurement solution could add to a toolkit of approaches to better validate source-specific greenhouse emissions inventories – an important new requirement of the UNFCCC COP21 (Paris) climate change agreement.

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Appendix A. Supplementary material

Supplementary data associated with this article can be found, in the online version, at https://doi.org/10.1016/j.wasman.2017.12.024.

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